

**PCT**WORLD INTELLECTUAL PROPERTY ORGANIZATION  
International Bureau

## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification <sup>6</sup> :</b> <b>C08K 3/00, C08J 5/18 // (C08K 3/00, 3:22, 3:34)</b>	<b>A1</b>	<b>(11) International Publication Number:</b> <b>WO 95/10563</b> <b>(43) International Publication Date:</b> 20 April 1995 (20.04.95)
<b>(21) International Application Number:</b> PCT/GB94/02165 <b>(22) International Filing Date:</b> 5 October 1994 <sup>5</sup> (05.10.94)  <b>(30) Priority Data:</b> 9321184.5                      14 October 1993 (14.10.93)                      GB  <b>(71) Applicant (for all designated States except US):</b> IMPERIAL CHEMICAL INDUSTRIES PLC [GB/GB]; Imperial Chemical House, Millbank, London SW1P 3JF (GB).  <b>(72) Inventors; and</b> <b>(75) Inventors/Applicants (for US only):</b> MILLS, Paul, David, Alan [GB/GB]; 16 Hillclose Avenue, Darlington, County Durham DL3 8BH (GB). OLEK, Anton, Richard [GB/GB]; 57 Farndale Drive, Guisborough, Cleveland TS14 8JJ (GB). RAKOS, Karl [GB/GB]; 11 Meadowdale, Chilton, Nr Ferryhill, County Durham DL17 0RW (GB).  <b>(74) Agents:</b> RHIND, John, Lessels et al.; ICI Materials, Intellectual Property Dept., P.O. Box 90, Wilton, Middlesbrough, Cleveland TS90 8JE (GB).		<b>(81) Designated States:</b> AU, BR, CA, CN, JP, KR, LT, US, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> POLYMERIC FILM  <b>(57) Abstract</b>  A mat polymeric film contains 74 to 99 % by weight of polyester, 1 to 20 % by weight of titanium dioxide particles having a volume distributed median particle diameter in the range from 0.1 to 2.0 $\mu\text{m}$ , and 0.1 to 6 % by weight of mica particles having a volume distributed median particle diameter in the range from 3 to 40 $\mu\text{m}$ . The polymeric film is suitable for use as a magnetic card.		

**FOR THE PURPOSES OF INFORMATION ONLY**

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT	Austria	GB	United Kingdom	MR	Mauritania
AU	Australia	GE	Georgia	MW	Malawi
BB	Barbados	GN	Guinea	NE	Niger
BE	Belgium	GR	Greece	NL	Netherlands
BF	Burkina Faso	HU	Hungary	NO	Norway
BG	Bulgaria	IE	Ireland	NZ	New Zealand
BJ	Benin	IT	Italy	PL	Poland
BR	Brazil	JP	Japan	PT	Portugal
BY	Belarus	KE	Kenya	RO	Romania
CA	Canada	KG	Kyrgyzstan	RU	Russian Federation
CF	Central African Republic	KP	Democratic People's Republic of Korea	SD	Sudan
CG	Congo	KR	Republic of Korea	SE	Sweden
CH	Switzerland	KZ	Kazakhstan	SI	Slovenia
CI	Côte d'Ivoire	LI	Liechtenstein	SK	Slovakia
CM	Cameroon	LK	Sri Lanka	SN	Senegal
CN	China	LU	Luxembourg	TD	Chad
CS	Czechoslovakia	LV	Latvia	TG	Togo
CZ	Czech Republic	MC	Monaco	TJ	Tajikistan
DE	Germany	MD	Republic of Moldova	TT	Trinidad and Tobago
DK	Denmark	MG	Madagascar	UA	Ukraine
ES	Spain	ML	Mali	US	United States of America
FI	Finland	MN	Mongolia	UZ	Uzbekistan
FR	France			VN	Viet Nam
GA	Gabon				

### Polymeric Film

This invention relates to a polymeric film and, in particular, to a matt polyester film.

5 Polymeric, and particularly polyester, films have been used in the production of a magnetic card, such as a credit card, and in particular a "pre-paid card", eg a telephone card, and an "intelligent card", which is, for example, capable of storing information relating to a number of financial transactions. A polyester film for use in a magnetic card is preferably opaque with the surface thereof exhibiting a matt finish. The matt finish is required to reduce specular light reflection from the surface of a magnetic card.  
10 Opaque polyester films have been traditionally produced by incorporating a white pigment, such as titanium dioxide into the film. Unfortunately, for certain applications the aforementioned polyester films may possess an unacceptable surface gloss.

During the normal film making process waste polyester film is recycled or reclaimed which can result in increased colour, particularly yellow, formation in the resultant film. Thus, there is also a commercial requirement for an opaque polyester film, especially for use in a magnetic card, to possess improved whiteness, particularly as regards reduced yellowness thereof.

We have now devised a polyester film suitable for use in a magnetic card which reduces or substantially overcomes at least one of the aforementioned problems.

20 Accordingly, the present invention provides a matt polymeric film comprising 74 to 99% by weight of polyester, 1 to 20% by weight of titanium dioxide particles having a volume distributed median particle diameter in the range from 0.1 to 2.0  $\mu\text{m}$ , and 0.1 to 6% by weight of mica particles having a volume distributed median particle diameter in the range from 3 to 40  $\mu\text{m}$ .

25 The invention also provides a method of producing a matt polymeric film comprising 74 to 99% by weight of polyester, 1 to 20% by weight of titanium dioxide particles having a volume distributed median particle diameter in the range from 0.1 to 2.0  $\mu\text{m}$ , and 0.1 to 6% by weight of mica particles having a volume distributed median particle diameter in the range from 3 to 40  $\mu\text{m}$ .

30 The invention further provides a magnetic card comprising a magnetic layer and a matt polymeric film which comprises 74 to 99% by weight of polyester, 1 to 20% by weight of titanium dioxide particles having a volume distributed median particle diameter in the range from 0.1 to 2.0  $\mu\text{m}$ , and 0.1 to 6% by weight of mica particles having a volume distributed median particle diameter in the range from 3 to 40  $\mu\text{m}$ .

35 The polymeric film is a self-supporting film, ie a self-supporting structure capable of independent existence in the absence of a supporting base.

The polymeric film according to the invention may be formed from any synthetic, film-forming polyester material. Suitable thermoplastics polyester materials include a synthetic linear polyester which may be obtained by condensing one or more dicarboxylic acids or their lower alkyl (up to 6 carbon atoms) diesters, eg terephthalic acid, isophthalic acid, phthalic acid, 2,5- 2,6- or 2,7-naphthalenedicarboxylic acid, succinic acid, sebacic acid, adipic acid, azelaic acid, 4,4'-diphenyldicarboxylic acid, hexahydroterephthalic acid or 1,2-bis-p-carboxyphenoxyethane (optionally with a monocarboxylic acid, such as pivalic acid) with one or more glycols, particularly aliphatic glycols, eg ethylene glycol, 1,3-propanediol, 1,4-butanediol, neopentyl glycol and 1,4-cyclohexanedimethanol. A polyethylene terephthalate or polyethylene naphthalate film is preferred. A polyethylene terephthalate film is particularly preferred, especially such a film which has been biaxially oriented by sequential stretching in two mutually perpendicular directions, typically at a temperature in the range 70 to 125°, and preferably heat set, typically at a temperature in the range 150 to 250°, for example as described in British patent 838708.

The amount of the polyester present in the polymeric film according to the invention is preferably in the range from 78 to 96% by weight, more preferably from 82.5 to 92%, and especially from 85 to 89.5% by weight based upon the total weight of the components present in the polymeric film.

The polymeric film according to the invention may be uniaxially oriented, but is preferably biaxially oriented by drawing in two mutually perpendicular directions in the plane of the film to achieve a satisfactory combination of mechanical and physical properties. Formation of the film may be effected by any process known in the art for producing an oriented polyester film, for example a tubular or flat film process.

In a tubular process simultaneous biaxial orientation may be effected by extruding a thermoplastics polyester tube which is subsequently quenched, reheated and then expanded by internal gas pressure to induce transverse orientation, and withdrawn at a rate which will induce longitudinal orientation.

In the preferred flat film process a film-forming polyester is extruded through a slot die and rapidly quenched upon a chilled casting drum to ensure that the polymer is quenched to the amorphous state. Orientation is then effected by stretching the quenched extrudate in at least one direction at a temperature above the glass transition temperature of the polyester. Sequential orientation may be effected by stretching a flat, quenched extrudate firstly in one direction, usually the longitudinal direction, ie the forward direction through the film stretching machine, and then in the transverse direction. Forward stretching of the extrudate is conveniently effected over a set of

rotating rolls or between two pairs of nip rolls, transverse stretching then being effected in a stenter apparatus. Stretching is effected to an extent determined by the nature of the film-forming polyester, for example a linear polyester is usually stretched so that the dimension of the oriented polyester film is from 2.5 to 4.5 times its original dimension in the, or each direction of stretching.

A stretched film may be, and preferably is, dimensionally stabilised by heat-setting under dimensional restraint at a temperature above the glass transition temperature of the film-forming polyester but below the melting temperature thereof, to induce crystallisation of the polyester.

The polymeric film according to the invention is preferably opaque and preferably exhibits a Transmission Optical Density (TOD) (Macbeth Densitometer; type TD 902; transmission mode) in the range from 0.4 to 1.75, more preferably from 0.6 to 1.6, especially from 0.8 to 1.5, and particularly from 0.9 to 1.4. The aforementioned TOD ranges are particularly applicable to a 190  $\mu\text{m}$  thick polymeric film.

The surface of a polymeric film according to the invention is matt and preferably exhibits a 60° gloss value, measured as herein described, of less than 60%, more preferably less than 50%, especially in the range from 5% to 40%, and particularly from 15% to 35%.

The polymeric film according to the invention is white and suitably exhibits a whiteness index, measured as herein described, of greater than 80, preferably greater than 85, more preferably greater 90, especially greater than 95, and particularly greater than 100 units.

The polymeric film according to the invention preferably exhibits a yellowness index, measured as herein described, of less than or equal to 3, more preferably less than or equal to 1, especially less than or equal to -1, and particularly less than or equal to -3.

The polymeric film according to the invention preferably exhibits a root mean square surface roughness ( $R_q$ ), measured as herein described, in the range from 200 to 1500 nm, more preferably from 400 to 1200 nm, and especially from 500 to 1000 nm. The polymeric film preferably has a mean surface slope ( $\Delta q$  - root mean square surface slope of the profile throughout the assessment length), measured as herein described, of greater than 2°, more preferably greater than 3°, and especially greater than 4°.

The individual or primary titanium dioxide particles suitably have a mean crystal size, as determined by electron microscopy, in the range from 0.05 to 0.4  $\mu\text{m}$ , preferably from 0.1 to 0.2  $\mu\text{m}$ , and more preferably of approximately 0.15  $\mu\text{m}$ . In a

preferred embodiment of the invention, the primary titanium dioxide particles aggregate to form clusters or agglomerates comprising a plurality of titanium dioxide particles. The aggregation process of the primary titanium dioxide particles may take place during the actual synthesis of the titanium dioxide and/or during the polyester and film making process.

5 The aggregated titanium dioxide filler preferably has a volume distributed median particle diameter (equivalent spherical diameter corresponding to 50% of the volume of all the particles, read on the cumulative distribution curve relating volume % to the diameter of the particles - often referred to as the "D(v,0.5)" value), as  
10 determined by laser diffraction, of from 0.3 to 1.5  $\mu\text{m}$ , more preferably from 0.4 to 1.2  $\mu\text{m}$ , and particularly from 0.5 to 0.9  $\mu\text{m}$ .

The size distribution of the titanium dioxide particles is also an important parameter, for example the presence of excessively large particles can result in the film exhibiting unsightly 'speckle', ie where the presence of filler aggregates in the film can be discerned with the naked eye. It is preferred that none of the titanium dioxide  
15 particles incorporated into the polymeric film should have an actual particle size exceeding 50  $\mu\text{m}$ . Particles exceeding such a size may be removed by sieving processes which are known in the art. However, sieving operations are not always totally successful in eliminating all particles greater than a chosen size. In practice,  
20 therefore, the size of 99.9% by number of the titanium dioxide particles should not exceed 50  $\mu\text{m}$ , preferably should not exceed 30  $\mu\text{m}$ , and more preferably should not exceed 20  $\mu\text{m}$ . Preferably at least 90%, more preferably at least 95% of the titanium dioxide filler particles are within the range of the volume distributed median particle diameter  $\pm 0.5 \mu\text{m}$ , and particularly  $\pm 0.3 \mu\text{m}$ .

25 The amount of titanium dioxide incorporated into the polymeric film desirably should be in the range from 3 to 17% by weight, more preferably from 6 to 14% by weight, and especially from 8 to 12% by weight based upon the total weight of the components present in the polymeric film.

30 The titanium dioxide particles may be of anatase or rutile crystal form. The titanium dioxide particles preferably comprise a major portion of anatase, more preferably at least 60% by weight, particularly at least 80%, and especially approximately 100% by weight of anatase. The particles can be prepared by standard procedures, such as using the chloride process or preferably by the sulphate process.

35 In one embodiment of the invention the titanium dioxide particles are coated preferably with inorganic oxides such as aluminium, silicon, zinc, magnesium or mixtures thereof. Preferably the coating additionally comprises an organic compound,

such as fatty acids and preferably alkanols, suitably having from 8 to 30, preferably from 12 to 24 carbon atoms. Polydiorganosiloxanes or polyorganohydrogensiloxanes, such as polydimethylsiloxane or polymethylhydrogensiloxane are suitable organic compounds.

5           The coating is applied to the titanium dioxide particles in aqueous suspension. The inorganic oxides are precipitated in aqueous suspension from water-soluble compounds such as sodium aluminate, aluminium sulphate, aluminium hydroxide, aluminium nitrate, silicic acid or sodium silicate.

10           The mica particles preferably have a volume distributed median particle diameter, as hereinbefore defined, as determined by laser diffraction, in the range from 8 to 30  $\mu\text{m}$ , more preferably in the range from 11 to 25  $\mu\text{m}$ , and especially from 14 to 20  $\mu\text{m}$ .

15           As regards particle size distribution, the mica particles preferably have a 90th percentile in the volume distributed cumulative particle size distribution of greater than 2  $\mu\text{m}$ , more preferably greater than 4  $\mu\text{m}$ , and especially greater than 5  $\mu\text{m}$ . In addition, the mica particles preferably have a 10th percentile in the volume distributed cumulative particle size distribution of less than 55  $\mu\text{m}$ , more preferably less than 40  $\mu\text{m}$ , and especially less than 30  $\mu\text{m}$ .

20           The ratio, of preferably at least 70%, more preferably at least 80%, and particularly at least 90% by number, of the maximum width to the minimum width of the mica particles is preferably less than 4:1, more preferably less than 2:1, and particularly less than 1.5:1.

25           In a preferred embodiment of the invention the mica particles suitably have a thickness in the range from approximately 0.1 to 0.6  $\mu\text{m}$ . The average thickness of the mica particles is preferably in the range from 0.2 to 0.5  $\mu\text{m}$ , more preferably from 0.3 to 0.4  $\mu\text{m}$ .

30           In order to obtain the advantageous properties of the present invention the concentration of mica particles present in the polymeric film should be in the range from 0.1 to 6.0% by weight, preferably 1.0 to 5.0% by weight, particularly 2.0 to 3.5% by weight, and especially 2.5 to 3.0% by weight, based upon the total weight of the components present in the polymeric film.

35           Particle size of the titanium dioxide and mica particles may be measured by electron microscope, coulter counter, sedimentation analysis and static or dynamic light scattering. Techniques based on laser light diffraction are preferred. The median particle size may be determined by plotting a cumulative distribution curve representing the percentage of particle volume below chosen particle sizes and measuring the 50th

percentile. The volume distributed median particle diameter of the filler particles is suitably measured using a Malvern Instruments Mastersizer MS 15 Particle Sizer after dispersing the filler in ethylene glycol in a high shear (eg Chemcoll) mixer. For non-spherical particles, such as planar particles like mica, a spherical average equivalent diameter is calculated.

The titanium dioxide and mica particles may be added to the polyester film or polyester film-forming material at any point in the film manufacturing process prior to the extrusion of the polyester. For example, the titanium dioxide and/or mica particles may be added during monomer transfer or in the autoclave, although it is preferred to incorporate the particles as a glycol dispersion during the esterification reaction stage of the polyester synthesis. Alternatively, the titanium dioxide and/or mica particles may be added as a dry powder into the polymer melt via a twin-screw extruder or by masterbatch technology.

The ratio by weight of titanium dioxide particles to mica particles present in a polymeric film according to the invention is preferably in the range from 10:1 to 1:1, more preferably from 6:1 to 2:1, and especially from 5:1 to 3:1.

In a preferred embodiment of the invention, the polymeric film comprises an optical brightener. An optical brightener may be included at any stage of the polymer or polymeric film production. For a polyester, it is preferred to add the optical brightener to the glycol, or alternatively by subsequent addition to the polyester prior to the formation of the polymeric film, eg by injection during extrusion. The optical brightener is preferably added in amounts of from 50 to 1500 ppm, more preferably from 200 to 1000 ppm, and especially from 400 to 600 ppm by weight based upon the weight of polymeric material in the film. Suitable optical brighteners include those available commercially under the trade names "Uvitex" MES, "Uvitex" OB, "Leucopur" EGM and "Eastobrite" OB-1.

The thickness of the polymeric film according to the invention is preferably in the range from 12 to 400  $\mu\text{m}$ , more preferably from 50 to 350  $\mu\text{m}$ , especially from 150 to 250  $\mu\text{m}$ , and particularly from 160 to 220  $\mu\text{m}$ .

A polymeric film according to the invention may be coated on one or both surfaces with one or more additional primer, coating, ink, lacquer, magnetic and/or metal layers, for example to form a laminate or composite which exhibits improved properties, such as antistatic, adhesion promoting or release, compared with the component materials.

A preferred adhesion promoting coating layer comprises an acrylic resin. The acrylic resin is preferably thermoset and preferably comprises at least one monomer



derived from an ester of acrylic acid and/or an ester of methacrylic acid, and/or derivatives thereof. In a preferred embodiment of the invention, the acrylic resin comprises greater than 50 mole %, preferably less than 98 mole %, more preferably from 60 to 97 mole %, especially from 70 to 96 mole %, and particularly from 80 to 94 mole % of at least one monomer derived from an ester of acrylic acid and/or an ester of methacrylic acid, and/or derivatives thereof. A preferred acrylic resin for use in the present invention preferably comprises an alkyl ester of acrylic and/or methacrylic acid where the alkyl group contains up to ten carbon atoms such as methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, terbutyl, hexyl, 2-ethylhexyl, heptyl, and n-octyl. Polymers derived from an alkyl acrylate, for example ethyl acrylate and butyl acrylate, together with an alkyl methacrylate are preferred. Polymers comprising ethyl acrylate and methyl methacrylate are particularly preferred. The acrylate monomer is preferably present in a proportion in the range 30 to 65 mole %, and the methacrylate monomer is preferably present in a proportion in the range of 20 to 60 mole %.

A preferred acrylic resin, derived from 3 monomers comprises 35 to 60 mole % of ethyl acrylate/ 30 to 55 mole % of methyl methacrylate/2 to 20 mole % of acrylamide or methacrylamide, and especially comprising approximate molar proportions 46/46/8% respectively of ethyl acrylate/methyl methacrylate/acrylamide or methacrylamide, the latter polymer being particularly effective when thermoset - for example, in the presence of about 25 weight % of a methylated melamine-formaldehyde resin.

A preferred acrylic resin, derived from 4 monomers comprises a copolymer comprising comonomers (a) 35 to 40 mole % alkyl acrylate, (b) 35 to 40 mole % alkyl methacrylate, (c) 10 to 15 mole % of a monomer containing a free carboxyl group, and (d) 15 to 20 mole % of a monomer containing sulphonic acid and/or a salt thereof.

Ethyl acrylate is a particularly preferred monomer (a) and methyl methacrylate is a particularly preferred monomer (b). Monomer (c) containing a free carboxyl group, ie a carboxyl group other than those involved in the polymerisation reaction by which the copolymer is formed, suitably comprises a copolymerisable unsaturated carboxylic acid, and is preferably selected from acrylic acid, methacrylic acid, maleic acid, and/or itaconic acid; with acrylic acid and itaconic acid being particularly preferred. The sulphonic acid monomer (d) is preferably aromatic and may be present as the free acid and/or a salt thereof, for example as the ammonium, substituted ammonium, or an alkali metal, such as lithium, sodium or potassium, salt. The sulphonate group does not participate in the polymerisation reaction by which the adherent copolymer resin is formed. The sulphonic acid monomer is preferably p-styrene sulphonic acid and/or a salt thereof.

Prior to the deposition of a coating medium onto the polymeric film according to the invention, the exposed surface thereof may, if desired, be subjected to a chemical or physical surface-modifying treatment to improve the bond between that surface and the subsequently applied coating layer. A preferred treatment is corona discharge, which may be effected in air at atmospheric pressure with conventional equipment using a high frequency, high voltage generator, preferably having a power output of from 1 to 20 kw at a potential of 1 to 100 kv. Discharge is conveniently accomplished by passing the film over a dielectric support roller at the discharge station at a linear speed preferably of 1.0 to 500 m per minute. The discharge electrodes may be positioned 0.1 to 10.0 mm from the moving film surface. Alternatively, the polymeric film surface may be pretreated with an agent known in the art to have a solvent or swelling action on the polymer layer. Examples of such agents which are particularly suitable for the treatment of a polyester film surface include a halogenated phenol dissolved in a common organic solvent, eg a solution of p-chloro-m-cresol, 2,4-dichlorophenol, 2,4,5- or 2,4,6- trichlorophenol or 4-chlororesorcinol in acetone or methanol.

The coating medium may be applied to an already oriented polymeric film, but application of the coating medium is preferably effected before or during the stretching operation.

In particular, it is preferred that the coating medium should be applied to the surface of a polymeric film between the two stages (longitudinal and transverse) of a thermoplastics film biaxial stretching operation. Such a sequence of stretching and coating is especially preferred for the production of a coated polymeric film comprising a linear polyester, which is preferably firstly stretched in the longitudinal direction over a series of rotating rollers, coated with the coating layer, and then stretched transversely in a stenter oven, preferably followed by heat setting.

In one embodiment of the invention the polymeric film additionally comprises a heat-sealable layer. The heat-sealable layer should be capable of forming a heat-seal bond to itself and/or to the substrate, by heating to soften the polymeric material of the heat-sealable layer and applying pressure without softening or melting the polyester material of the substrate layer.

The heat-sealable layer suitably comprises a polyester resin, particularly a copolyester resin derived from one or more dibasic aromatic carboxylic acids, such as terephthalic acid, isophthalic acid and hexahydroterephthalic acid, and one or more glycols, such as ethylene glycol, diethylene glycol, triethylene glycol and neopentyl glycol. Typical copolyesters which provide satisfactory heat-sealable properties are

those of ethylene terephthalate and ethylene isophthalate, especially in the molar ratios of from 50 to 90 mole % ethylene terephthalate and correspondingly from 50 to 10 mole % ethylene isophthalate. Preferred copolyesters comprise from 65 to 85 mole % ethylene terephthalate and from 35 to 15 mole % ethylene isophthalate, and especially a copolyester of about 82 mole % ethylene terephthalate and about 18 mole % ethylene isophthalate.

A polymeric film according to the invention, particularly when coated with a heat-sealable layer, may be used to produce credit or identification cards. In particular, the heat-sealable layer may be printed thereon with ink or dye from conventional printing sources, for example in a thermal transfer printing process. The printed film may be heat-sealed to another polymeric film, preferably a polyester, more preferably a transparent film which can act as a protective cover for the printed layer.

A magnetic card, as hereinbefore described, preferably comprises the following layers, in order, (i) a magnetic layer, for storing the relevant information, (ii) a polymeric film layer, according to the present invention, and (iii) a graphics layer or printing layer, which provides visual information, such as the type of card, owner etc. A range of conventional binders may be used for the magnetic coating materials, which are well-known to the man skilled in the art. A wide range of inks and lacquers may be used in the graphics layer, such as aqueous and organic solvent-based materials, particularly electron beam- and UV-curable inks.

Polymeric films according to the invention may be used in other applications where a white opaque film exhibiting a matt surface is required, such as in photographic applications, non-magnetic information storage and as writing materials.

In this specification the following test methods have been used to determine certain properties of the polymeric film:

(i) Transmission Optical Density (TOD)

TOD of the film was measured using a Macbeth Densitometer TD 902 (obtained from Dent and Woods Ltd, Basingstoke, UK) in transmission mode.

(ii) Matt Surface Property

The 60° gloss value of the film surface was measured using a Dr Lange Reflectometer RB3 (obtained from Dr Bruno Lange, GmbH, Dusseldorf, Germany) based on the principles described in ASTM D 523.

(iii) Whiteness Index and Yellowness Index

The whiteness index and yellowness index of the film was measured using a Colorgard System 2000, Model/45 (manufactured by Pacific Scientific) based on the principles described in ASTM D 313.

(iv) Surface Roughness

The film surface root mean square roughness ( $R_q$ ) and mean surface slope ( $\Delta q$ ) were measured using a Rank Taylor-Hobson Talysurf 10 (Leicester, UK) employing a cut-off length of 0.25 mm.

5 The invention is illustrated by reference to the following examples.

Example 1

Polyethylene terephthalate polymer comprising 10% by weight of anatase titanium dioxide having a volume distributed median particle diameter of 0.7  $\mu\text{m}$ , 3.5% by weight of mica (SX300 Mica, supplied by Microfine Minerals Ltd, Derby, UK) of  
10 volume distributed median particle diameter of 18  $\mu\text{m}$ , and 460 ppm of optical brightener (Eastobrite OB1, Eastman Kodak), was extruded through a film-forming die onto a water cooled rotating, quenching drum to yield an amorphous cast composite extrudate. The cast extrudate was heated to a temperature of about 80°C and then stretched longitudinally at a forward draw ratio of 3.2:1. The polymeric film was passed  
15 into a stenter oven, where the film was stretched in the sideways direction to approximately 3.4 times its original dimensions. The biaxially stretched polymeric film was heat set at a temperature of about 225°C. Final film thickness was approximately 188  $\mu\text{m}$ .

20 The polymeric film was subjected to the test procedures described herein and exhibited the following properties.

- (i) Transmission Optical Density (TOD) = 1.0
- (ii) 60° gloss value = 31%
- (iii) Whiteness Index = 98 units  
Yellowness Index = -3.0 units
- 25 (iv) Root mean square roughness ( $R_q$ ) = 570 nm  
Mean surface slope ( $\Delta q$ ) = 4.3°

Example 2

The procedure of Example 1 was repeated except that the optical brightener was omitted. The polymeric film was subjected to the test procedures described herein and  
30 exhibited the following properties.

- (i) Transmission Optical Density (TOD) = 1.0
- (ii) 60° gloss value = 31%
- (iii) Whiteness Index = 84 units  
Yellowness Index = 1.5 units
- 35 (iv) Root mean square roughness ( $R_q$ ) = 590 nm  
Mean surface slope ( $\Delta q$ ) = 4.4°

**Example 3**

This is a comparative example not according to the invention. The procedure of Example 1 was repeated except that the mica was omitted. The polymeric film was subjected to the test procedures described herein and exhibited the following properties.

- 5 (i) Transmission Optical Density (TOD) = 0.9
- (ii) 60° gloss value = 74%
- (iii) Whiteness Index = 98 units  
Yellowness Index = -3.0 units
- 10 (iv) Root mean square roughness (Rq) = 80 nm  
Mean surface slope ( $\Delta q$ ) = 1.6°

The above examples illustrate the improved properties of polymeric films according to the present invention.

Claims

1. A matt polymeric film comprising 74 to 99% by weight of polyester, 1 to 20% by weight of titanium dioxide particles having a volume distributed median particle diameter in the range from 0.1 to 2.0  $\mu\text{m}$ , and 0.1 to 6% by weight of mica particles having a volume distributed median particle diameter in the range from 3 to 40  $\mu\text{m}$ .
2. A polymeric film according to claim 1 wherein the film additionally comprises 50 to 1500 ppm of an optical brightener.
3. A polymeric film according to either one of claims 1 and 2 wherein the Transmission Optical Density (TOD) of the film is in the range from 0.6 to 1.6.
4. A polymeric film according to any one of the preceding claims wherein the surface of the film has a 60° gloss value of less than 60%.
5. A polymeric film according to any one of the preceding claims wherein the film comprises 6 to 14% by weight of titanium dioxide particles.
6. A polymeric film according to any one of the preceding claims wherein the film comprises 1 to 5% by weight of mica particles.
7. A polymeric film according to any one of the preceding claims wherein the volume distributed median particle diameter of mica particles is in the range from 11 to 25  $\mu\text{m}$ .
8. A polymeric film according to any one of the preceding claims wherein the titanium dioxide particles are approximately 100% by weight anatase.
9. A method of producing a matt polymeric film comprising 74 to 99% by weight of polyester, 1 to 20% by weight of titanium dioxide particles having a volume distributed median particle diameter in the range from 0.1 to 2.0  $\mu\text{m}$ , and 0.1 to 6% by weight of mica particles having a volume distributed median particle diameter in the range from 3 to 40  $\mu\text{m}$ .
10. A magnetic card comprising a magnetic layer and a polymeric film according to any one of claims 1 to 8, or produced according to claim 9.

## INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB 94/02165

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C08K3/00 C08J5/18 //(C08K3/00,3:22,3:34)

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08K C08J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DATABASE WPI Derwent Publications Ltd., London, GB; AN 86-030467 & ES,A,8 506 366 (GANDIA M T) 1 November 1985 see abstract ---	1
A	EP,A,0 043 063 (LETRON GMBH) 6 January 1982 see page 5, paragraph 2 ---	1
A	US,A,4 910 235 (SATAKE ET AL) 20 March 1990 see example 1 -----	1

☐ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

## \* Special categories of cited documents :

- \*A\* document defining the general state of the art which is not considered to be of particular relevance
- \*E\* earlier document but published on or after the international filing date
- \*L\* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- \*O\* document referring to an oral disclosure, use, exhibition or other means
- \*P\* document published prior to the international filing date but later than the priority date claimed

\*T\* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

\*X\* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

\*Y\* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

\*&\* document member of the same patent family

Date of the actual completion of the international search

21 December 1994

Date of mailing of the international search report

10. 01. 95

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax (+31-70) 340-3016

Authorized officer

Siemens, T

## INTERNATIONAL SEARCH REPORT

ation on patent family members

nal Application No

PCT/GB 94/02165

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP-A-0043063	06-01-82	DE-A- 3024582 JP-A- 57030576	21-01-82 18-02-82
US-A-4910235	20-03-90	JP-A- 63289030	25-11-88